

Application No. 10/617,852
Reply to Office Action of September 23, 2004

Amendments to the Specification

Please replace Paragraph [0017] with the following amended paragraph:

[0017] In one embodiment the process of the present invention is for the manufacture of a silicoaluminophosphate molecular sieve of framework type AEL, wherein the molar ratio of organic template to ~~Al₂O₃~~ Al₂O₃ in the synthesis mixture is less than 3.

Please replace Paragraph [0018] with the following amended paragraph:

[0018] In a further embodiment the process of the present invention is for the manufacture of a silicoaluminophosphate molecular sieve of framework type CHA, wherein the molar ratio of organic template to ~~Al₂O₃~~ Al₂O₃ is 2 or greater, more preferably 3 or greater.

Please replace Paragraph [0019] with the following amended paragraph:

[0019] In a preferred embodiment the ~~P₂O₅/Al₂O₃~~ P₂O₅/Al₂O₃ ratio in the synthesis mixture is within the range 0.8 to 1.3.

Please replace Paragraph [0023] with the following amended paragraph:

[0023] In a further embodiment of the present invention there is provided a silicoaluminophosphate molecular sieve, substantially of AEL framework type, comprising within its intra-crystalline structure at least one template which contains one or more tertiary dialkylbutylamines, wherein the alkyl groups are not butyl. Preferable, the tertiary dialkylbutylamine is ~~simethylbutylamine~~ dimethylbutylamine.

Application No. 10/617,852

Reply to Office Action of September 23, 2004

Please replace Paragraph [0026] with the following amended paragraph:

[0026] In ~~yeta~~ yet a further embodiment the present invention provides for the use of a template comprising at least one compound which contains one or more tertiary dialkylbutylamines, wherein the alkyl groups are not butyl, in the synthesis of silicoaluminophosphates of the AEL or CHA framework type.

Please replace Paragraph [0039] with the following amended paragraph:

[0039] The single ~~C₄H₉~~ C₄H₉ group in formula (I) may be n-butyl or iso-butyl or mixtures thereof, preferably n-butyl.

Please replace Paragraph [0040] with the following amended paragraph:

[0040] The sources of aluminum, phosphorus and silicon suitable for use in the synthesis of molecular sieves according to the present invention are typically those known in the art or as described in the literature for the production of the SAPO concerned. The aluminum source may be, for example, an aluminum oxide (alumina), optionally hydrated, an aluminum salt, especially a phosphate, an aluminate, or a mixture thereof. A preferred source is a hydrated alumina, most preferably pseudoboehmite, which contains about ~~75% Al₂O₃ and 25% H₂O~~ 75% Al₂O₃ and 25% H₂O by weight. Advantageously, the source of phosphorus is a phosphoric acid, especially orthophosphoric acid, but other sources, for example, organic phosphates, e.g., triethyl phosphate, and aluminophosphates may be used. Advantageously, the source of silicon is silica, for example colloidal silica, fumed silica, or an organic silicon source, e.g., a tetraalkyl orthosilicate, especially tetraethyl orthosilicate.

Application No. 10/617,852

Reply to Office Action of September 23, 2004

Please replace Paragraph [0041] with the following amended paragraph:

[0041] In the process of the present invention selection of the correct molar ratio of organic template to Al_2O_3 is important in determining which silicoaluminophosphate framework type is crystallized. Accordingly if the desired silicoaluminophosphate is of framework type AEL then the molar ratio of organic template to Al_2O_3 should be less than 3 and preferably is 2.5 or less, more preferably 2.0 or less. If the desired silicoaluminophosphate is of framework type CHA then the molar ratio of organic template to Al_2O_3 should be 2 or more, more preferably 3 or more. In a preferred embodiment the $\text{P}_2\text{O}_5/\text{Al}_2\text{O}_3$ ratio in the synthesis mixture is within the range 0.8 to 1.3.

Please replace Paragraph [0045] with the following amended paragraph:

[0045] Aluminium chlorohydrol, a hydroxylated aluminium based sol containing a chloride counter ion, has the general formula of $\text{Al}_m\text{O}_n(\text{OH})_p\text{Cl}_q \cdot x(\text{H}_2\text{O})$ wherein m is 1 to 20, n is 1 to 8, o is 5 to 40, p is 2 to 15, and x is 0 to 30. In one embodiment, the binder is $\text{Al}_{13}\text{O}_4(\text{OH})_{24}\text{Cl}_7 \cdot 12(\text{H}_2\text{O})$ as is described in G.M. Wolterman, et al., Stud. Surf. Sci. and Catal., 76, pages 105-144 (1993), which is herein incorporated by reference. In another embodiment, one or more binders are combined with one or more other non-limiting examples of alumina materials such as aluminium oxyhydroxide, γ -alumina, boehmite, diaspore, and transitional aluminas such as α -alumina, β -alumina, γ -alumina, δ -alumina, ϵ -alumina, κ -alumina, and ρ -alumina, aluminium trihydroxide, such as gibbsite, bayerite, nordstrandite, doyleite, and mixtures thereof.

Application No. 10/617,852

Reply to Office Action of September 23, 2004

Please replace Paragraph [0083] with the following amended paragraph:

[0083] In one embodiment of the disengaging system, the disengaging system includes a disengaging vessel; typically a lower portion of the disengaging vessel is a stripping zone. In the stripping zone the coked molecular sieve catalyst composition is contacted with a gas, preferably one or a combination of steam, methane, carbon dioxide, carbon monoxide, hydrogen, or an inert gas such as argon, preferably steam, to recover adsorbed hydrocarbons from the coked

molecular sieve catalyst composition that is then introduced to the regeneration system. In another embodiment, the stripping zone is in a separate vessel from the disengaging vessel and the gas is passed at a gas hourly superficial velocity (GHSV) of ~~from 1 hr⁻¹ to about 20,000 hr⁻¹~~ from 1 hr⁻¹ to about 20,000 hr⁻¹ based on the volume of gas to volume of coked molecular sieve catalyst composition, preferably at an elevated temperature from 250°C to about 750°C, preferably from about 350°C to 650°C, over the coked molecular sieve catalyst composition.

Please replace Paragraph [0087] with the following amended paragraph:

[0087] Typically, the WHSV ranges ~~from about 1 hr⁻¹ to about 5000 hr⁻¹, preferably from about 2 hr⁻¹ to about 3000 hr⁻¹, more preferably from about 5 hr⁻¹ to about 1500 hr⁻¹, and most preferably from about 10 hr⁻¹ to about 1000 hr⁻¹~~ from about 1 hr⁻¹ to about 5000 hr⁻¹, preferably from about 2 hr⁻¹ to about 3000 hr⁻¹, more preferably from about 5 hr⁻¹ to about 1500 hr⁻¹, and most preferably from about 10 hr⁻¹ to about 1000 hr⁻¹. In one preferred embodiment, the WHSV is greater than ~~20 hr⁻¹~~ 20 hr⁻¹; preferably the WHSV for conversion of a feedstock containing methanol and dimethyl ether is in the range of ~~from about 20 hr⁻¹ to about 300 hr⁻¹~~ from about 20 hr⁻¹ to about 300 hr⁻¹.

Application No. 10/617,852

Reply to Office Action of September 23, 2004

Please replace Paragraph [0089] with the following amended paragraph:

[0089] In one preferred embodiment of the process for converting an oxygenate to olefin(s) using a silicoaluminophosphate molecular sieve catalyst composition, the process is operated at a WHSV of at least ~~20 hr⁻¹~~ 20 hr⁻¹ and a Temperature Corrected Normalized Methane Selectivity (TCNMS) of less than 0.016, preferably less than or equal to 0.01. See for example U.S. Patent No. 5,952,538, which is herein fully incorporated by reference.

Please replace Paragraph [0090] with the following amended paragraph:

[0090] In another embodiment of the processes for converting an oxygenate such as methanol to one or more olefin(s) using a molecular sieve catalyst composition, the WHSV is ~~from 0.01 hr⁻¹ to about 100 hr⁻¹~~ from 0.01 hr⁻¹ to about 100 hr⁻¹, at a temperature of from about 350°C to 550°C, and silica to Me₂O₃ (Me is a Group IIIA or VIII element from the Periodic Table of Elements) molar ratio of from 300 to 2500. See for example EP-0 642 485 B1, which is herein fully incorporated by reference.

Please replace Paragraph [0093] with the following amended paragraph:

[0093] Non-limiting examples of the regeneration medium include one or more of oxygen, ~~O₃, SO₃, N₂O, NO, NO₂, N₂O₅~~ O₃, SO₃, N₂O, NO, NO₂, N₂O₅, air, air diluted with nitrogen or carbon dioxide, oxygen and water (U.S. Patent No. 6,245,703), carbon monoxide and/or hydrogen. The regeneration conditions are those capable of burning coke from the coked catalyst composition, preferably to a level less than 0.5 weight percent based on the total weight of the coked molecular sieve

Application No. 10/617,852

Reply to Office Action of September 23, 2004

catalyst composition entering the regeneration system. The coked molecular sieve catalyst composition withdrawn from the regenerator forms a regenerated molecular sieve catalyst composition.

Please replace Paragraph [0110] with the following amended paragraph:

[0110] The following ingredients are mixed, in sequence, and blended into a uniform gel using a microhomogenizer (Tissue Tearor Model 98730 available from Biospec Products Inc, USA): 7.04g 85 wt% ~~H₃PO₄~~ H₃PO₄ (obtained from Aldrich Chemical Company), 13.4g ~~H₂O~~ H₂O, 0.53g Cabosil™ (available from Cabot Corporation, Illinois, USA), 3.66g Catapal™ A (71.5 wt% ~~Al₂O₃~~ Al₂O₃, available from CONDEA Vista Company, Texas, USA), and then 5.3g of N, N-dimethylbutylamine (DMBA) (obtained from Aldrich Chemical Company, USA). The molar ratio of the ingredients is as follows:

2.0 DMBA:1.0 Al₂O₃:0.3 SiO₂:1.15 P₂O₅:34 H₂O

Please replace Paragraph [0111] with the following amended paragraph:

[0111] The gel was then divided into two equal parts and placed into a Parr bomb with Teflon liner, and heated to 170°C for 10 days with no agitation. The solid product was centrifuged and washed several times with deionized water until the conductivity of the supernatant was below 50 mS/cm, and was then dried in a 60 °C vacuum oven overnight to provide a silicoaluminophosphate of the AEL framework type, comprising N,N-dimethylbutylamine within its intra-crystalline structure. X-ray powder patterns of the product confirmed that the product is a silicoaluminophosphate of the AEL framework type. The solid product yield is 15.0% after 3 days of crystallization, based on the weight of the starting synthesis mixture. Elemental analysis of the products gives: Al, 19.7%; P, 21.6%; Si, 1.58% which corresponds to a molar composition ~~Al_{11.0}Si_{0.077}P_{0.955}~~ Al_{1.0}Si_{0.077}P_{0.955}.

Application No. 10/617,852

Reply to Office Action of September 23, 2004

Please replace Paragraph [0113] with the following amended paragraph:

[0113] The procedure of Example 1 was repeated to prepare 175g of gel. Mixing in this case was achieved using a simple blender. The gel was then transferred to a stainless steel autoclave equipped with a stirrer and was sealed before being heated to 170°C for 40 hours during which the gel was stirred at 240 rpm. The solid product was centrifuged and washed several times with deionized water until the conductivity of the supernatant was below 50 mS/cm, and was then dried in a 60 °C vacuum oven overnight to provide a silicoaluminophosphate of the AEL framework type, comprising N,N-dimethylbutylamine within its intra-crystalline structure. X-ray powder patterns of the product confirmed that the product is a silicoaluminophosphate of the AEL framework type. The solid product yield is 20.9% after 3 days of crystallization, based on the weight of the starting synthesis mixture. Elemental analysis of the products gives: Al, 18.4%; P, 20.7%; Si, 2.06% which corresponds to a molar composition $Al_{1.0}Si_{0.108}P_{0.980}$.